In Operando Thermal Diagnostics of Electrochemical Cells

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Project ID: bat427

Background & Relevance

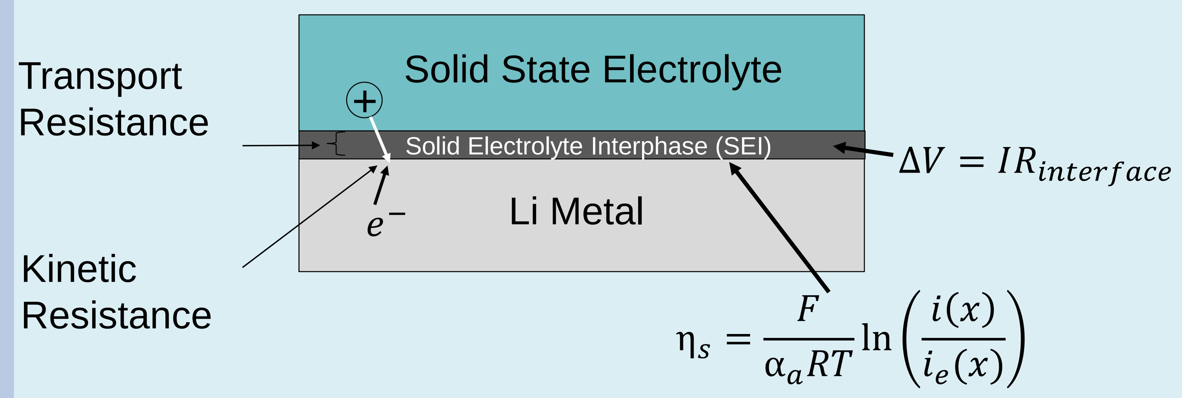
Diagnostics of interface processes through thermal signatures will improve…

1) Safety: Operando detection of dendrite and interface morphology change

2) Performance: Understanding factors contributing to interface related transport and kinetic overpotential

Timeline
Start:10/01/2019
End: 09/30/2023
50% Complete

Budget
Total: $840k
FY2022:$300k



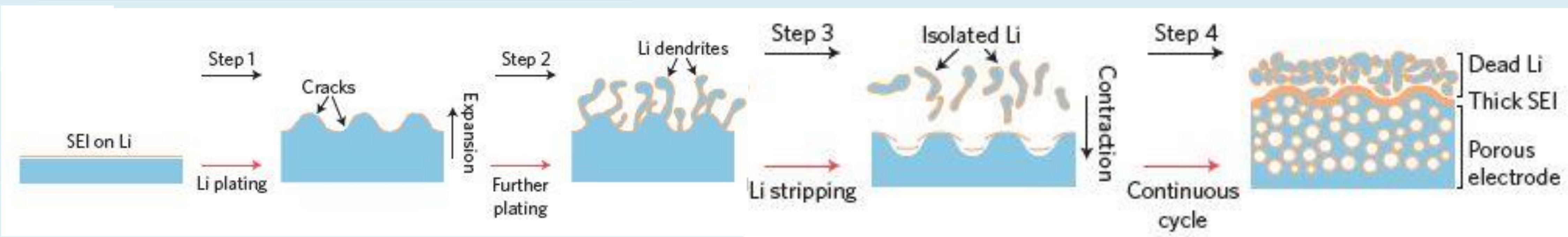
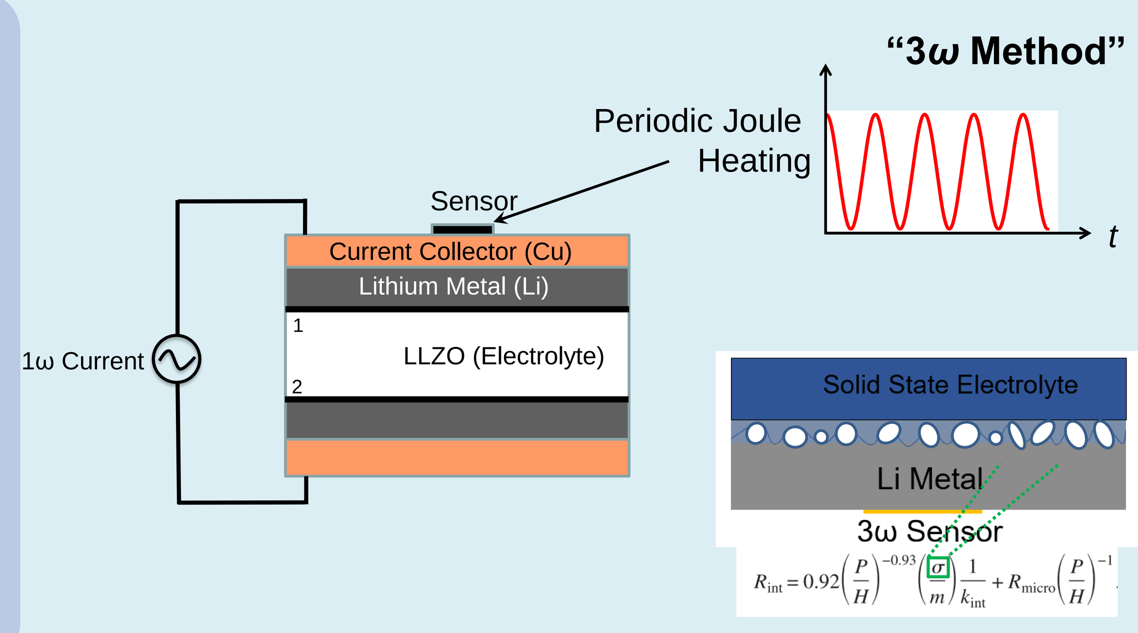
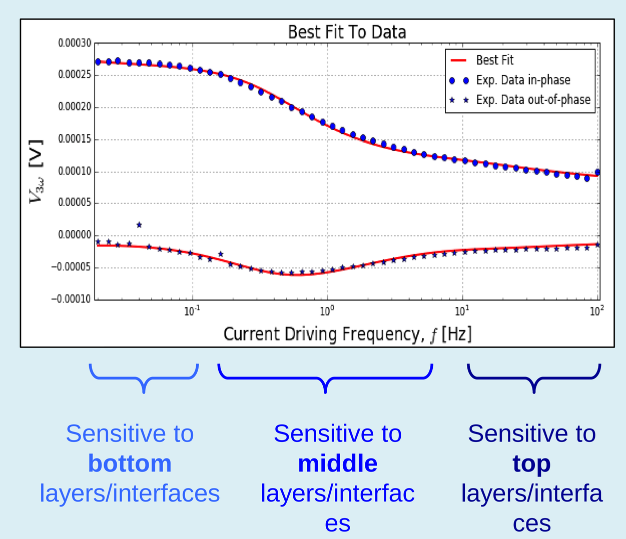


Image from D. Lin *et al.*, "Reviving the lithium metal anode for high-energy batteries," *Nature Nanotechnology*, vol. **12**, pp. 194-206, 2017.

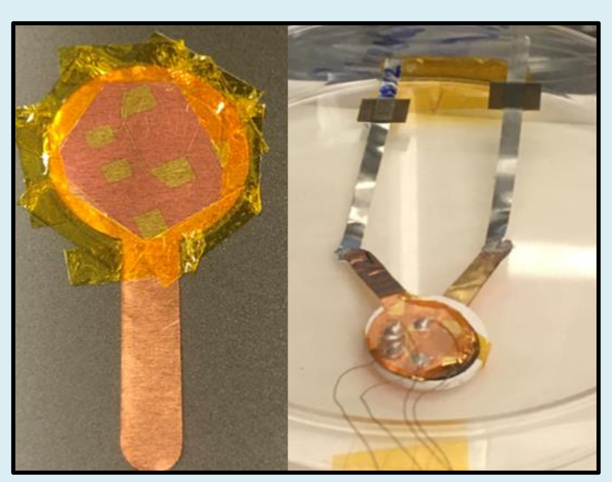
Approach: Thermal Wave Sensing

- Relate Interface Morphology with the thermal interface resistance
- Extract Electrochemical Information from heat generation signature

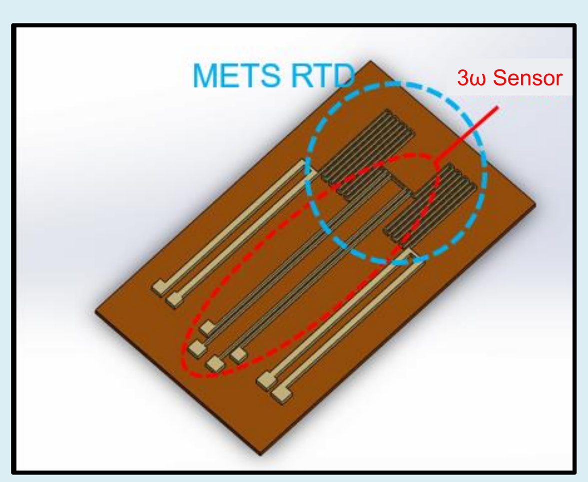




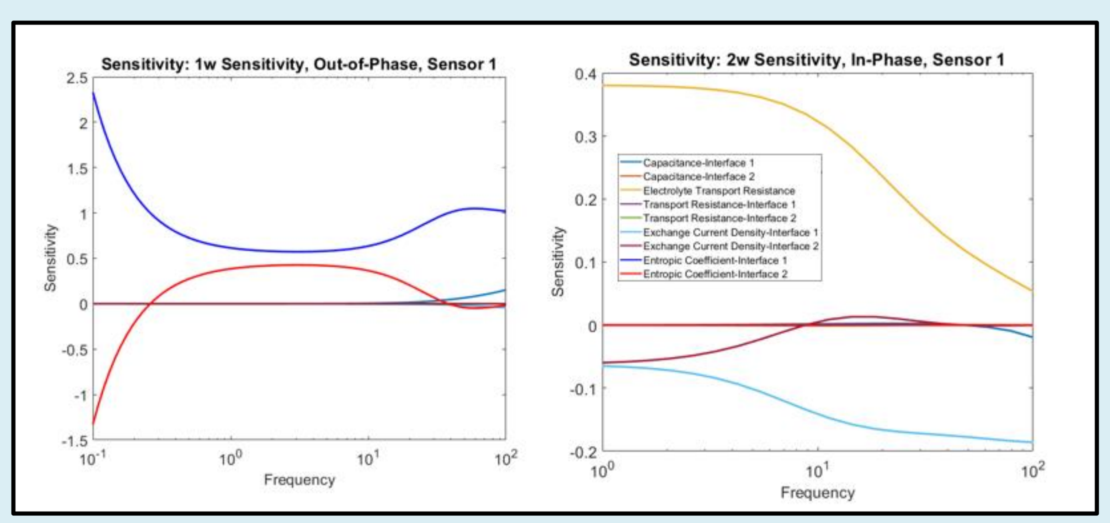
Multi-harmonic ElectroThermal Spectroscopy (METS)



Cell assembly with a 3ω Sensor



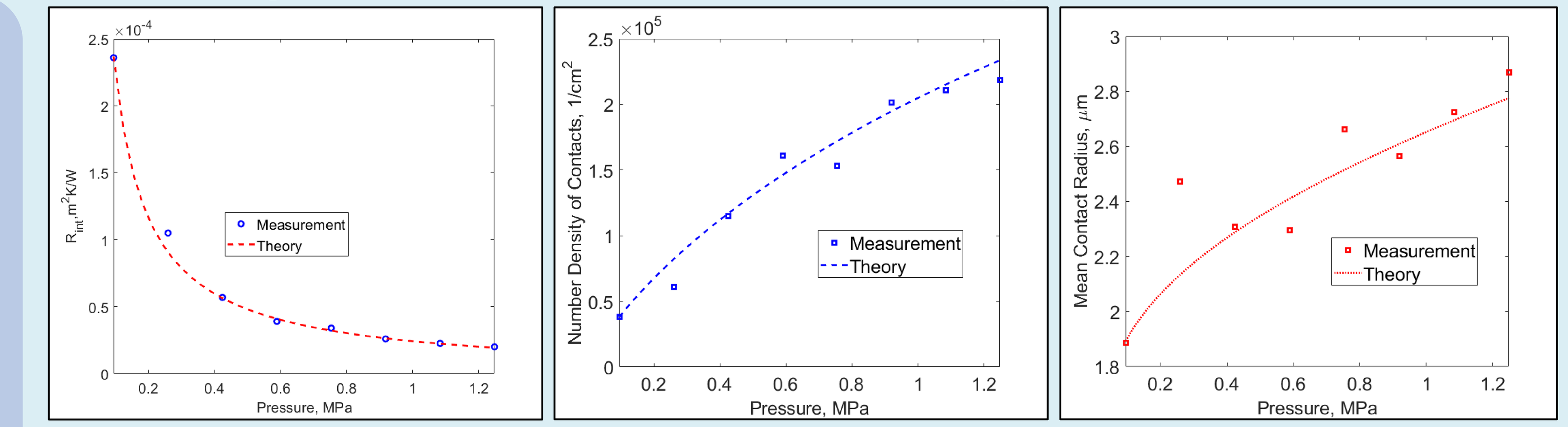
Resistance Sensor for METS



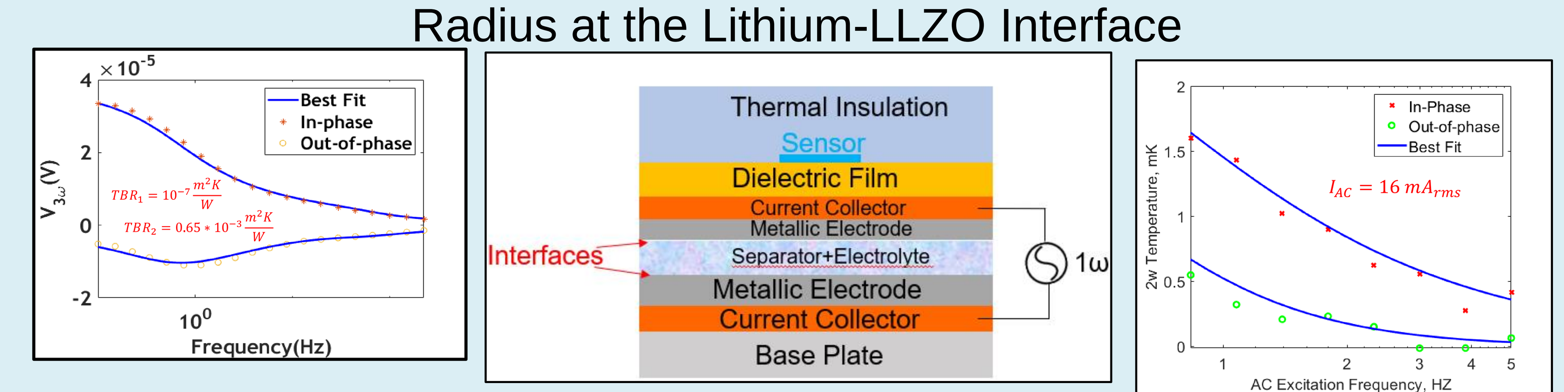
Electrochemical Properties from METS

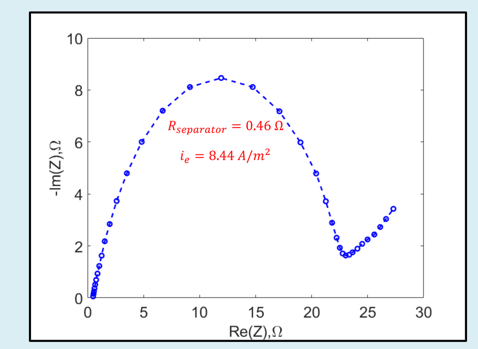
Technical Accomplishments & Progress

- Measured thermal interface resistance of Lithium-LLZO interface evolution as a function of pressure and cycles
- Related the thermal interface resistance with interface morphology
- Verified Multi-harmonic ElectroThermal Spectroscopy (METS) for a model system

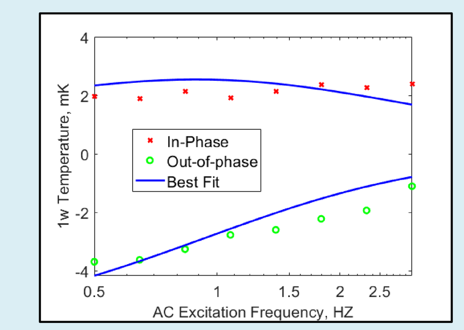


Measurement of Thermal Contact Resistance, Contact Density and Contact Radius at the Lithium-LLZO Interface





Quantity	Value	Method
Exchange Current Density (i_e)	8.82 A/m^2	2w METS
	8.44 A/m^2	EIS
Electrolyte Resistance (R_t)	0.46 Ω	EIS
SEI Resistance (R_{SEI})	0.1 Ω	2w METS
Entropic Coefficient ($\frac{dU}{dT}$)	0.53 $\frac{mV}{K}$	1w METS



Verification of METS for a model system

Summary & Future Outlook

Summary

1) Thermal Wave Sensing allows non-invasive, operando measurement of interface properties. By using the 3ω method to measure the thermal interface resistance measurement, we have non-invasively extracted properties related to the interface morphology.

2) Heat generation signature at the different harmonics and frequency allows separation of entropic, kinetic and transport properties with spatial resolution. We have established a new electrochemical method: Multi-harmonic ElectroThermal Spectroscopy (METS) based on this principle and verified it on a model electrochemical system.

Quarter	Milestones & Go/No-Go	Status
Q3, FY21	Sensitivity analysis and sample design optimization to maximize measurement sensitivity to target electrochemical properties.	Completed
Q4, FY21	Baseline impedance spectroscopy of cells for ion and electron mobility.	Completed
Q1, FY22	Identification of theoretical model relating the interface morphology to thermal contact resistance	Completed
Q2, FY22	Measured change to TCR/morphology with cell cycling correlated with EIS.	Completed

Remaining Challenges & Future Work

1) METS measurement on Li-LLZO system to isolate the contribution of the transport and kinetic overpotential at the lithium metal-LLZO interface.

2) Ex situ characterization of interface morphology to validate the theory relating the morphology to TCR